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(57) **Abstract**
SUBJECT The particle diameter in the end of dried powder obtained is uniform, and generation of fines is controlled, and the granulation method of CMC which can obtain the carboxymethylcellulose ether alkali salt (the following "CMC" is called) which was moreover excellent in handlability with high bulk density is provided.
Means for Solution By cracking massive CMC in hydrous isopropyl alcohol, granular CMC is corned in liquid.

Claim(s)
Claim 1A granulation method of carboxymethylcellulose ether alkali salt corning in liquid by cracking massive carboxymethylcellulose ether alkali salt in hydrous isopropyl alcohol.
Claim 2 In a system which consists of hydrous isopropyl alcohol containing massive carboxymethylcellulose ether alkali salt at the time of corning in the above-mentioned liquid, A

content ratio of carboxymethylcellulose ether alkali salt 5 to 30% of the weight of the whole system. A granulation method of the carboxymethylcellulose ether alkali salt according to claim 1 in which a content ratio of isopropyl alcohol is set to 55 to 80% of the weight of the whole system, and a moisture content ratio is set to 15 to 40% of the weight of the whole system.
Claim 3 Reynolds number (Re) computed by the following formula (1) showing the flowability of a hydrous isopropyl alcohol solution system containing massive carboxymethylcellulose ether alkali salt in a crack in the above-mentioned liquid, A granulation method of the carboxymethylcellulose ether alkali salt according to claim 1 or 2 which is 1000 or more.
Equation 1 Re=(D-U-rho)/mu ... (1)
In a formula (1), as for shuttlecock peripheral speed (cm/sec) and rho of D, liquid density (g/cm^3) and mu are a shuttlecock diameter (cm) and U liquid viscosity (g/cm-sec).
Claim 4 Granular carboxymethylcellulose ether alkali salt whose bulk density the whole contains **particles of within the limits with a particle diameter of 149-2000 micrometers** 80% of the weight or more, and is 0.4g/ml or more.

Detailed Description of the Invention
0001
Field of the InventionA granulation method of the carboxymethylcellulose ether alkali salt (the following "CMC" is called) in which this invention is characterized by processing among liquid, The purpose of being related with granular CMC which is a granulation article, and preventing preservation of the work environment by the prevention from raising dust, automation of the taking-out ON at the time of a transfer, and adhesion in a container on granular material management of CMC, The solubility to water is improved, and it excels in fastmelt, and is related with the granulation method of granular CMC which can aim at improvement in the handlability in various fields, and CMC.
0002
Description of the Prior Art From the former, CMC used for a various application uses pulp etc. as a raw material, performs an etherification reaction after an alkali-cellulose-ized reaction, and subsequently, deliquoring filtration is neutralized and carried out with acetic acid etc., and it produces crude CMC. Then, water content adds the methanol aqueous solution below 30 % of the weight (it omits the following "%") to this crude CMC, and performs demineralization refining to it. It is manufactured by carrying out hot air drying of the filtered refining CMC, and next, grinding it.
0003
Problem(s) to be Solved by the Invention However, CMC obtained by the above-mentioned process has a problem shown below.
0004** Since many fibrous things contain with fines (particle diameter of 75 micrometers or less) unevenly **the particle diameter of obtained CMC** , there is much dusting and there is a problem in workability.
** When using CMC of the above-mentioned fines, it is generated by dust, and it is ****** about an adverse effect to work environment.
** Since unevenness and fibrous fines have much particle diameter of obtained CMC, in the dissolution to water, unmixed-in lump of flour generates and, moreover, the dissolution takes a long time bad to solubility.
** Since CMC is fines, it is easy to absorb moisture, solidify with product pressure, and handlability gets worse.
0005 Since it has the above problems, mobility is good and granulation-ization of miniaturized CMC is considered. For example, as a granulation method of CMC, in the manufacturing process of CMC, the solvent used for this reaction is separated after the reaction of CMC, and the so-called crude CMC of the shape of friable wet is produced. And 1-2-times the amount water is sprayed on this crude CMC to CMC under stirring mixing, and CMC which melted a part of fibrous CMC, and hardened and granulation(keratinization)-ized it is produced. Then, CMC granulation-ized **above** is immersed in a lot of methanol for the purpose of drying, and after separating this methanol and drying, the granulation method of CMC of obtaining granular CMC which dries and serves as a granulation article is raised (US,2715124,B). However, in the above-mentioned granulation method, since gel CMC by spraying of water is uneven as mentioned above, there is a problem that a granulation article with a particle size uniform as a

product is not obtained. In order for gel CMC to stick and to grow up granular as mentioned above, the grain shape state is formed in the particles by which uneven shape was formed in the surface like konpeito, and, as a result, has the problem that mobility worsens. CMC of the degree of high ether substitution has high water solubility, and since it is easy to dissolve, adhesion to a device is strong, and since CMC sticks further and it does not become particles, there is a problem of being unable to carry out **granulation** -izing. The yield of product CMC is bad and the above-mentioned granulation method is a high cost at the whole.

0006 On the other hand, applicant of this application has proposed the method of carrying out spray drying and granulation-izing by making it flow down on a rotating disc, and making the solvent-water content slurry of CMC atomize previously (Tokuganhei6-215058). However, the CMC granulation article obtained by this method had low bulk density, and it turned out that it is not desirable in respect of the mobility of CMC, and miniaturization.

0007 It was made in view of such a situation, the particle diameter in the end of dried powder obtained is uniform, and generation of fines is controlled, the granulation method of CMC which can obtain CMC which was moreover excellent in handlability with high bulk density is carried out, and this invention sets offer of granular CMC as the purpose.

0008

Means for Solving the Problem In order to attain the above-mentioned purpose, this invention makes the 1st gist a granulation method of CMC corned in liquid by cracking massive CMC in hydrous isopropyl alcohol, The whole contains not less than 80%, and particles of within the limits with a particle diameter of 149-2000 micrometers make the 2nd gist granular CMC whose bulk density is 0.4g/ml or more.

0009

Embodiment of the Invention Regardless of the degree of ether substitution, this invention from the degree of low ether substitution to the degree of high ether substitution, That is, even if it is water-soluble high CMC, in order to obtain the granulation article of quality CMC, it is the method of corning in liquid by cracking massive CMC in isopropyl alcohol (the following "IPA" is called) of water content. The system which consists of a hydrous IPA solution which contains massive CMC used as a raw material in the granulation in this liquid, The CMC granulation article with high bulk density (granular CMC) in which the content ratio of CMC is uniform particle diameter when the content ratio of IPA sets to 55 to 80% of the whole system and a moisture content ratio sets to 15 to 40% of the whole system 5 to 30% of the whole system, and generation of fines was controlled is obtained, and it is more effective. In the describing **above** granulation method in liquid, granular CMC which is uniform particle diameter and by which generation of fines of 75 micrometers or less was controlled for particle diameter is obtained by performing the Reynolds number (Re) which shows the flowability of the hydrous IPA solution system containing massive CMC under the conditions set or more to 1000. Therefore, when dissolving obtained granular CMC in water, the generation phenomenon of the unmixed-in lump of flour seen conventionally does not arise, but it dissolves in water promptly. As a result, the dissolving time at the time of use by a various application is shortened substantially, and since there is little dusting and mobility is still better, it excels also in handlability. At the time of granular CMC use, there is little generating of dust and it does not cause aggravation of work environment.

0010 And that to which not less than 80% of all the particles existed in within the limits with a particle diameter of 149-2000 micrometers, and bulk density was set **ml** in 0.4g /or more as granular CMC is excellent in mobility and solubility, and does not produce a raising dust problem, either, but, moreover, comes to have good handlability.

0011 Below, this invention is explained in detail.

0012 Although the degrees of average ether substitution are 0.4-3.0 and the target CMC can be applied to any CMC in the granulation method of CMC of this invention in that it is applicable also to CMC which has a binding property, In particular, in the high degree of average ether substitution of hydrophilic nature, CMC of 1.5-3.0 serves as a suitable object. Although sodium salt, potassium salt, ammonium salt, etc. are raised as a kind of alkali salt, it is usually sodium salt. CMC may be any of refining CMC obtained via the process of removing a byproduction salt using crude CMC and this crude CMC. Since the refining effects are inferior when demineralization refining is carried out after granulation-izing by this invention, it is preferred to use refining CMC.

0013 The granulation method of CMC of this invention is performed as follows, for example. That is, pulp which is a publicly known method is conventionally used for a raw material, neutralization processing is performed via an alkali-cellulose-ized reaction and an etherification

reaction, and crude CMC is prepared by carrying out deliquoring filtration. On the other hand, prepare the partially aromatic solvent (hydrous IPA) which mixed IPA with water by the weight ratio within the limits of water/IPA=40 / 60 - 70/30, and the above-mentioned crude CMC is received, The weight of the above-mentioned crude CMC throws in 5-20 times the amount of this partially aromatic solvent, it stirs at 30-75 ** for 0.5 to 1 hour, and a demineralization step is performed (desalting process). And it deliquors with a centrifuge, a decanter, a screw-type continuation liquid separation machine, a filter, etc., and wet-like refining CMC is produced. Refining CMC produced here is mass material in which CMC solid content has elasticity **like the shape of the shape of agar to konnyaku which has a binding property by IPA consisting of 15 to 35% of presentation 45 to 65%** whose water is 20 to 40%. Thus, massive refining CMC obtained serves as high water content of 45 to 65% in the above-mentioned desalting process.

0014 Next, supply massive refining CMC and IPA (solvent) of 20% or less of water content to a predetermined mixer by the shape of wet acquired above, the moisture which remains in refining CMC is made to shift to it into the solvent which is IPA, and dehydrating treatment of refining CMC is performed. While performing this dehydrating treatment, by carrying out shuttlecock stirring and cracking massive refining CMC within a mixer, it cuts finely and the granulation in liquid is performed. Since said refining CMC is high water content, IPA of 20% or less of water content which uses by the above-mentioned granulation in liquid is used in order to adjust the water content to 5 to 30% of range. That is, IPA of 20% or less of water content is a useful solvent which can prevent re-binding at the time of a granulation, and can perform dehydrating treatment of refining CMC simultaneously. And since use of methanol and ethanol which are, other organic solvents, for example, lower alcohol, other than IPA, has the high water solubility and adhesion of CMC, it becomes impossible to dissolve and to perform the granulation in liquid, and it is not preferred. Since a bad smell cannot deaerate use of butanol easily strongly, it becomes expensive economically and is not preferred.

0015 Next, CMC corned among liquid removes a solvent with a centrifuge, then performs desiccation for 5 to 60 minutes at 60-110 ** using a fluidized-drying machine or a decompression type dryer. Thus, granular CMC is obtained.

0016 In the above-mentioned granulation in liquid, a mixer type mixer as shown in drawing 1 is raised as a mixer which supplies refining CMC and hydrous IPA. That is, the granulation in liquid is performed, rotating the knife type agitating blades 1, cutting massive refining CMC2 small, and adding rotation in this mixer, in the hydrous IPA solution 3 stirred with predetermined Reynolds number.

0017 In the above-mentioned granulation in liquid, it is preferred to perform the Reynolds number (Re) of the value which shows the flowability of hydrous IPA solution 3 system containing massive refining CMC2 by 1000 or more strong shuttlecock stirring. Reynolds number (Re) is 4000-30000 especially preferably. That is, granular CMC which is uniform particle diameter and by which generation of fines of 75 micrometers or less was controlled for particle diameter comes to be obtained by setting Reynolds number (Re) or more to 1000. The above-mentioned Reynolds number (Re) is computed by the formula (1) shown below.

0018

Equation 2 Re=(D-U-rho)/mu ... (1)

In a formula (1), as for shuttlecock peripheral speed (cm/sec) and rho of D, liquid density (g/cm^3) and mu are a shuttlecock diameter (cm) and U liquid viscosity (g/cm-sec).

0019 In the above-mentioned granulation in liquid, the number of rotations at the time of stirring is suitably set up within the limits of 100-2000 rpm according to the massive amount of refining CMC etc. to supply. As for the temperature of massive refining CMC to supply, mixing time is set as 10-50 ** in 5 to 30 minutes.

0020 And on the occasion of the granulation in liquid, supply of massive refining CMC may be supplied continuously and may be added to a batch process. It may add at once by a batch type, and the amount of supply is not limited.

0021 The granulation morphosis in such a granulation stage in liquid is explained in detail.

0022 That is, CMC after an ether reaction at the time of crude CMC manufacture is maintaining fibrous voice, and its density with bulky is low. A low water solvent usually performs demineralization refining in order to remove impurities, such as a byproduction salt contained in this crude CMC. However, in this invention, it precedes supplying the granulation in liquid as mentioned above, and demineralization refining is carried out beforehand in high hydrous IPA whose water content is 40 to 70%. If a demineralization step is performed and filtrate

separation is carried out in this high hydrous IPA, massive refining CMC of the shape of wet which contained moisture 45 to 65% will be obtained. At this process, since fibrous CMC contains moisture, it is welded and changes from the shape of agar to a massive konnyaku-like solid. The good state of dispersibility is held by stirring, without this solid dissolving in an IPA system solvent.

0023 Subsequently, after carrying out filtrate separation, for the purpose of drying of a granulation in liquid, and massive refining CMC, konnyaku-like massive refining CMC is supplied with 20% or less of hydrous IPA (solvent), rotates agitating blades and carries out stirring mixing here. And massive refining CMC is promptly cracked in liquid, and granulation cutting is carried out at particle diameter of about 1 mm. Here, drying by moisture shift to IPA advances, and, simultaneously with a crack, spherical particles are obtained by rotation.

0024 Thus, not less than 80% of the whole particle has a particle size within limits which are the particle diameter of 149-2000 micrometers, and obtained granular CMC does not contain fines with a particle diameter of 75 micrometers or less. Or even if it contains fines with a particle diameter of 75 micrometers or less, they are very little content of a grade which does not cause a soluble fall by an adverse effect by dust, and unmixed-in-lump-of-flour generation. Measurement of the above-mentioned particle diameter is measured by standard sieve (indicated to JIS Z8801), and can check the whole particle size distribution. Bulk density which is 0.4-0.8g/ml 0.4g/ml or more has **especially granular CMC that comprises such particle size distribution** still higher bulk density.

0025 Below, it combines with a comparative example and working example is described.

0026 First, three kinds of refining CMC which serves as a raw material on the occasion of a granulation of this invention and from which the degree of etherification (DS) differs were produced.

0027Production of refining CMC by solvent method** Stock pulpIt ground to 0.3 mm in diameter, having applied **the Kohjin Co., Ltd. make and NDSP (the sulfate method pulp)** to a grinder, and pulp of 5% of moisture content was obtained. then, the 30-l. reaction vessel made from SUS filled up with hydrous IPA which consists of IPA20 weight section (it omits the following "part") and two copies of water -- 0.8 copy of this crushed pulp -- in addition, it stirred for about 20 minutes at 20 **. And an alkali-cellulose-ized reaction was performed by in addition stirring 1.1 copies of flake articles of caustic alkali of sodium for 60 minutes at 30 ** for 5 minutes 100% to this. Next, 2.4 copies of monochloroacetic acid content IPA solutions were added over 10 minutes 50% to this. Then, after carrying out temperature up to 70 ** over about 30 minutes, again, it cooled to 30 ** and 1.1 copies of flake articles of caustic alkali of sodium were added over 5 minutes 100%, and 2nd addition was performed these 30 minutes afterward, having covered 2.4 copies of monochrome acetic acid content IPA solutions 50% for 10 minutes. Then, temperature up was carried out to 70 ** over 30 minutes, and an etherification reaction for 60 minutes was completed in this 70 ** state. After an etherification reaction, it cooled to 40 ** over 10 minutes, 0.3 copy of acetic acid solution was added 50%, and superfluous caustic alkali of sodium in a system was neutralized. And deliquoring filtration of this was carried out with a centrifuge, and 8.7 copies of crude CMC (sodium salt) of the degree 2.1 of etherification (DS) were produced. Water of CMC was **IPA of a presentation of this crude CMC** 28.5% 19.5% 52%.

0028 Six copies of water and a 60% hydrous solvent of four copies of IPA were added to one copy of above-mentioned crude CMC, stirring was performed to it for 60 minutes at 50 **, and a demineralization step was performed to it. Then, deliquoring filtration was carried out with a centrifuge and 0.8 copy of wet-like massive refining CMC (sodium salt) was obtained. Thus, obtained refining CMC was mass material which consists of 30% of solid content, 48% of water, and IPA22% of presentation, and has agar-like softness and brittleness.

0029Production of refining CMC by solvent method** Stock pulpIt ground to 0.3 mm in diameter, having applied **the Kohjin Co., Ltd. make and NDSP (the sulfate method pulp)** to a grinder, and pulp of 5% of moisture content was obtained. Then, 1.0 copy of this crushed pulp was added to the 30-l. reaction vessel made from SUS filled up with hydrous IPA which consists of 25 copies of IPA, and 2.5 copies of water, and it stirred for about 20 minutes at 20 **. And an alkali-cellulose-ized reaction was performed by in addition stirring 1.0 copy of caustic soda aqueous solution for 60 minutes at 30 ** for 5 minutes 40% to this. Next, 0.78 copy of monochloroacetic acid content IPA solution was added over 5 minutes 50% to this. Then, temperature up was carried out to 70 ** over about 30 minutes. An etherification reaction for 90 minutes was performed in this 70 ** state. After an etherification reaction, it cooled to 40 ** over 10 minutes, 0.3 copy of acetic acid solution was added 50%, and

superfluous caustic alkali of sodium in a system was neutralized. And deliquoring filtration of this was carried out with a centrifuge, and 2.5 copies of crude CMC (sodium salt) of the degree 0.6 of etherification (DS) were produced. Water of CMC was **IPA of a presentation of this crude CMC** 23.6% 11.4% 65%.

0030 Seven copies of water and a 70% hydrous solvent of three copies of IPA were added to one copy of above-mentioned crude CMC, stirring was performed to it for 30 minutes at 50 **, and a demineralization step was performed to it. Then, deliquoring filtration was carried out with a centrifuge and 2.3 copies of wet-like massive refining CMC (sodium salt) were obtained. Thus, obtained refining CMC was mass material which consists of 21% of solid content, 57% of water, and IPA22% of presentation, and has konnyaku-like elasticity and brittleness.

0031Production of refining CMC by solvent method** Stock pulpIt ground to 0.3 mm in diameter, having applied **the Kohjin Co., Ltd. make and NDSP (the sulfate method pulp)** to a grinder, and pulp of 5% of moisture content was obtained. Then, 1.0 copy of this crushed pulp was added to the 30-l. reaction vessel made from SUS filled up with hydrous IPA which consists of 25 copies of IPA, and 2.5 copies of water, and it stirred for about 20 minutes at 20 **. And an alkali-cellulose-ized reaction was performed by in addition stirring 2.7 copies of caustic soda aqueous solutions for 60 minutes at 30 ** for 5 minutes 40% to this. Next, 2.3 copies of monochloroacetic acid content IPA solutions were added over 5 minutes 50% to this. Then, temperature up was carried out to 70 ** over about 30 minutes. An etherification reaction for 90 minutes was performed in this 70 ** state. After an etherification reaction, it cooled to 40 ** over 10 minutes, 0.3 copy of acetic acid solution was added 50%, and superfluous caustic alkali of sodium in a system was neutralized. And deliquoring filtration of this was carried out with a centrifuge, and 5.6 copies of crude CMC (sodium salt) of the degree 1.5 of etherification (DS) were produced. Water of CMC was **IPA of a presentation of this crude CMC** 27.3% 14.7% 58%.

0032 Nine copies of water and a 60% hydrous solvent of six copies of IPA were added to one copy of above-mentioned crude CMC, stirring was performed to it for 30 minutes at 50 **, and a demineralization step was performed to it. Then, deliquoring filtration was carried out with a centrifuge and 1.4 copies of wet-like massive refining CMC (sodium salt) were obtained. Thus, obtained refining CMC was mass material which consists of 26% of solid content, 50% of water, and IPA24% of presentation, and has konnyaku-like elasticity and brittleness.

0033

Working example 1-15, comparison working example 1-6 A granulation by this invention was performed as follows using refining CMC** produced as mentioned above, **, and **. That is, one copy of solid content of massive refining CMC shown in the after-mentioned table 1 and this 8-times the amount 10% hydrous IPA were added to this using a large-sized mixer type mixer (made by a domestic centrifuge company) with a capacity of 10 l. first provided with the knife type agitating blades 1 as shown in drawing 1. Each content ratio of CMC of the whole system at this time, IPA, and moisture is shown in the after-mentioned table 1. And stirring was started at a room temperature (25 **), and it stirred by setting promptly Reynolds number which shows a flow of a solution as a value shown in the after-mentioned table 2. Granulation conditions by the above-mentioned mixer are shown in the after-mentioned table 2. The above-mentioned Reynolds number is the value computed by said formula (1), combines with the after-mentioned table 2 each value of each factor and shuttlecock diameter D (cm), the shuttlecock peripheral speed U (cm/sec), the liquid density rho (g/cm^3), and the liquid viscosity mu (g/cm-sec) needed for calculation of Reynolds number, and shows it.

0034

Table 1

For drawings please refer to the original document.

0035

Table 2

For drawings please refer to the original document.

0036 While massive refining CMC was small cracked in hydrous IPA liquid as a result of the above-mentioned stirring, since it rotated in liquid, granular CMC in which the granule child was

formed spherically was obtained. The optical microscope photograph of granular CMC of one working example is shown in drawing 2. Thus, as for it, granular CMC obtained by the granulation in liquid turns out that the whole is approximately spherical. A lot of moisture contained in massive refining CMC shifted into IPA liquid on the occasion of the above-mentioned stirring. As a result, keratinization advanced with drying and, as for the CMC surface by which the granulation was carried out, solidification and the rise of density occurred. For this reason, CMC particles did not adhere easily to devices due to the shape of a pan pan, and the granulation article of good CMC was obtained. And the above-mentioned granulation process is performed in 5 to 20 minutes, and can be performed by any method of continuous system or a batch type.

0037 After the above-mentioned granulation, with the centrifuge, after carrying out deliquoring separation, the drying-by-warm-air machine performed desiccation of 2 hours at 100 ** warm air. Thus, granular CMC was obtained.

0038

Comparative examples 1-4 10 times the amount of hydrous methanol was added to this 20% using crude CMC** from which the degree of etherification produced in the manufacturing process of the above-mentioned refining CMC** - ** differs - **, demineralization washing was performed 2 times, and refining CMC was obtained. And in order to double a moisture content with above-mentioned working example for the above-mentioned refining CMC, spray water, it was made to increase and an 8-times the amount 10% hydrous methanol system performed granulation-ization after that on the conditions shown in the after-mentioned table 3 using the same mixer as above-mentioned working example (comparative examples 1-3). On the other hand, after hot air drying equipment performed desiccation of 100 **x 2 hours, the shock type pulverizing mill (made by Hosokawa Micron CORP.) ground deliquored refining CMC, and granular CMC was obtained (comparative example 4).

0039

Table 3

For drawings please refer to the original document.

0040

Conventional example 15 times the amount of hydrous methanol was added to this 20% using crude CMC** of the degree 1.5 of etherification produced in the manufacturing process of the above-mentioned refining CMC**, demineralization washing was performed 2 times, and refining CMC was obtained (CMC in refining CMC: 65%, 5% of moisture, methanol 30%). And in order to double a moisture content with above-mentioned working example for this refining CMC, sprayed water, it was made to increase to 50% of a moisture content, and CMC which melted a part of fibrous CMC, and hardened and (keratinization) corned it was produced. Then, for the purpose of drying, 10-times the amount 100%IPA was added to this CMC, and after separating the solvent and drying, granular CMC was obtained by performing 60 ** hot-air-drying processing. The optical microscope photograph of granular CMC of the obtained conventional example article is shown in drawing 3. Thus, as for obtained granular CMC, uneven shape was formed in the whole surface **like konpeito** the shape of whose a grain shape is.

0041 Thus, in each granular CMC of elegance, the yield and water content of the state of granular CMC corned under the granulation in liquid and obtained granular CMC are shown in the after-mentioned Table 4 and 5 the obtained working example article, a comparison working example article, a comparative example article, and conventionally. The yield of the above-mentioned granular CMC is computed by the following formula.

0042

Equation 3 Yield (%) = **(purity of granular CMC) /(purity of refining CMC)** x100 **0043**The bulk density of each obtained granular CMC, the content ratio of the particles of within the limits with a particle diameter **to the whole granular CMC** of 149-2000 micrometers, and the content ratio of a particle with a particle diameter of 75 micrometers or less are measured by a standard sieve (JIS Z8801), and the result is shown in the following Table 4 and 5.

0044

Table 4

For drawings please refer to the original document.

0045
Table 5

For drawings please refer to the original document.

0046 The chart figure showing the particle size distribution by measurement by the standard sieve (JIS Z8801) of five working example (the degree 1.5 of ether substitution) and nine working example (the degree 2.1 of ether substitution) is shown in drawing 4 (five working example) and drawing 5 (nine working example) out of measurement of the above-mentioned call counter, respectively, The numerical value acquired by the measurement is shown in the following Table 6 (five working example) and 7 (nine working example). 381.52 micrometers and the mean particle diameter of nine working example of the mean particle diameter of five working example by the above-mentioned call counter measurement were 814.41 micrometers, standard deviation was 597.28 micrometers, and standard deviation was 267.09 micrometers.
0047
Table 6

For drawings please refer to the original document.

0048
Table 7

For drawings please refer to the original document.

0049 The solubility of each CMC and the dissolution rate, the dusting characteristics, the amount of raising dust, adhesion, and mobility which were obtained were measured and evaluated in accordance with the following method. The result is shown in the after-mentioned table 8.
0050The solubility and the dissolution rate of granular CMC 800 ml of water was put into a 1000-ml beaker, and 8 g (1% concentration) of CMC samples were added into this. As a result, the thing of x and the above-mentioned middle evaluation of what became a letter of distribution immediately and was able to perform the aqueous solution quickly was displayed as **. **O and the thing which the unmixed-in-lump-of-flour-like lump was formed immediately and the aqueous solution took the long time** Time until it stirs lightly and a CMC sample dissolves was measured.
0051Dusting characteristics of CMC 1 / 2 capacity restoration of the CMC sample were carried out at the 100-ml screw pipe, and this was stirred up and down. As a result, the thing of x and the above-mentioned middle evaluation of a thing without ****** of fines was displayed as **. **what has many ****** of O and fines**
0052The amount of raising dust of CMC After putting 10g of CMC samples into the 300-ml Mayer flask and shaking at 10 cm of upper and lower sides 3 times, the floating CMC dust was absorbed by the suction collector which set the extraction thimble, and the amount of raising dust was measured.
0053Adhesion of CMC After filling up a 1-l. polyethylene bag with 200g of CMC samples and neglecting it on the 1st, the bag mouth was placed upside down, the natural fall of the CMC sample was carried out, and it took out from the inside of a bag. And the CMC powder quantity which adhered and remained in the bag was measured, and deposit efficiency was computed.
0054Mobility of CMC From the 20-cm-high place, it let the funnel whose inside diameter of the exit lower part is 10 mm pass, and the natural fall of 50 g of the CMC samples was carried out on the glass plate. And the diameter of CMC which fell and spread in the round form on the glass plate was measured. It can be said that what has a large diameter naturally has high mobility.
0055
Table 8

For drawings please refer to the original document.

0056 From the result of the above-mentioned table 8, a comparative example article is inferior to solubility, and its amount of raising dust is also high. It turns out that it is inferior also to mobility by high adhesion. On the other hand, the comparison working example article excels the comparative example article in each characteristic. The working example article produced by specifying Reynolds number or more as 1000 is dramatically excellent in solubility and mobility.
And it is clear that there are not the amount of raising dust and coating weight, and the good result was obtained in each characteristic.

0057
Effect of the Invention As mentioned above, in this invention, massive CMC is cracked all over hydrous IPA.
Therefore, it is the method of corning in liquid.
In the granulation method in this liquid, since it is not a method by spraying of water like before, for example even if it uses CMC whose hygroscopicity is the water-soluble high, large degree of high ether substitution as a raw material, granular CMC of the uniform particle size instead of particles in which unevenness by dissolution adhesion was formed can be manufactured easily. The particle diameter of granular CMC obtained is uniform, and generation of fines CMC of fibrous CMC with a particle diameter of 75 micrometers or less is controlled.
0058 And the system which consists of a hydrous IPA solution which contains massive CMC used as a raw material in the granulation in this liquid, Granular CMC with high bulk density by which the content ratio of CMC is uniform particle diameter when the content ratio of IPA sets to 70 to 90% of the whole system and a moisture content ratio sets to 5 to 40% of the whole system 5 to 25% of the whole system, and generation of fines was controlled is obtained, and it is effective. Granular CMC by which the Reynolds number (Re) which shows the flowability of the hydrous IPA solution system which contains massive CMC for the above-mentioned granulation in liquid is uniform particle diameter by carrying out under 1000 or more strong stirring conditions, and generation of fines of 75 micrometers or less was controlled for particle diameter is obtained. Therefore, when dissolving obtained granular CMC in water, the generation phenomenon of the unmixed-in lump of flour seen conventionally does not arise, but it dissolves in water promptly. As a result, large shortening of the dissolving time at the time of granular CMC use is realized, and since mobility is still better, it excels also in handlability. There is little generating of the dust in the time of use of granular CMC, and improvement in work environment is achieved. And since obtained granular CMC has good mobility, automation of constant feeding is attained when using it, the automation by production processes, such as paper and pulp industry and food stuff industry, is attained, and it is preferred.
0059 And as granular CMC, not less than 80% of all the particles exist in within the limits with a particle diameter of 149-2000 micrometers, and a thing of 0.4g/ml or more excels **bulk density** in mobility and solubility, and a raising dust problem is not produced, either, but, moreover, it comes to have good handlability.

Field of the InventionA granulation method of the carboxymethylcellulose ether alkali salt (the following "CMC" is called) in which this invention is characterized by processing among liquid, The purpose of being related with granular CMC which is a granulation article, and preventing preservation of the work environment by the prevention from raising dust, automation of the taking-out ON at the time of a transfer, and adhesion in a container on granular material management of CMC, The solubility to water is improved, and it excels in fastmelt, and is related with the granulation method of granular CMC which can aim at improvement in the handlability in various fields, and CMC.

Description of the Prior Art From the former, CMC used for a various application uses pulp etc. as a raw material, performs an etherification reaction after an alkali-cellulose-ized reaction, and subsequently, deliquoring filtration is neutralized and carried out with acetic acid etc., and it produces crude CMC. Then, water content adds the methanol aqueous solution below 30 % of the weight (it omits the following "%") to this crude CMC, and performs demineralization

refining to it. It is manufactured by carrying out hot air drying of the filtered refining CMC, and next, grinding it.

Effect of the Invention As mentioned above, in this invention, massive CMC is cracked all over hydrous IPA.
Therefore, it is the method of corning in liquid.
In the granulation method in this liquid, since it is not a method by spraying of water like before, for example even if it uses CMC whose hygroscopicity is the water-soluble high, large degree of high ether substitution as a raw material, granular CMC of the uniform particle size instead of particles in which unevenness by dissolution adhesion was formed can be manufactured easily. The particle diameter of granular CMC obtained is uniform, and generation of fines CMC of fibrous CMC with a particle diameter of 75 micrometers or less is controlled.
0058 And the system which consists of a hydrous IPA solution which contains massive CMC used as a raw material in the granulation in this liquid, Granular CMC with high bulk density by which the content ratio of CMC is uniform particle diameter when the content ratio of IPA sets to 70 to 90% of the whole system and a moisture content ratio sets to 5 to 40% of the whole system 5 to 25% of the whole system, and generation of fines was controlled is obtained, and it is effective. Granular CMC by which the Reynolds number (Re) which shows the flowability of the hydrous IPA solution system which contains massive CMC for the above-mentioned granulation in liquid is uniform particle diameter by carrying out under 1000 or more strong stirring conditions, and generation of fines of 75 micrometers or less was controlled for particle diameter is obtained. Therefore, when dissolving obtained granular CMC in water, the generation phenomenon of the unmixed-in lump of flour seen conventionally does not arise, but it dissolves in water promptly. As a result, large shortening of the dissolving time at the time of granular CMC use is realized, and since mobility is still better, it excels also in handlability. There is little generating of the dust in the time of use of granular CMC, and improvement in work environment is achieved. And since obtained granular CMC has good mobility, automation of constant feeding is attained when using it, the automation by production processes, such as paper and pulp industry and food stuff industry, is attained, and it is preferred.
0059 And as granular CMC, not less than 80% of all the particles exist in within the limits with a particle diameter of 149-2000 micrometers, and a thing of 0.4g/ml or more excels **bulk density** in mobility and solubility, and a raising dust problem is not produced, either, but, moreover, it comes to have good handlability.

Problem(s) to be Solved by the Invention However, CMC obtained by the above-mentioned process has a problem shown below.
0004** Since many fibrous things contain with fines (particle diameter of 75 micrometers or less) unevenly **the particle diameter of obtained CMC** , there is much dusting and there is a problem in workability.
** When using CMC of the above-mentioned fines, it is generated by dust, and it is ****** about an adverse effect to work environment.
** Since unevenness and fibrous fines have much particle diameter of obtained CMC, in the dissolution to water, unmixed-in lump of flour generates and, moreover, the dissolution takes a long time bad to solubility.
** Since CMC is fines, it is easy to absorb moisture, solidify with product pressure, and handlability gets worse.
0005 Since it has the above problems, mobility is good and granulation-ization of miniaturized CMC is considered. For example, as a granulation method of CMC, in the manufacturing process of CMC, the solvent used for this reaction is separated after the reaction of CMC, and the so-called crude CMC of the shape of friable wet is produced. And 1-2-times the amount water is sprayed on this crude CMC to CMC under stirring mixing, and CMC which melted a part of fibrous CMC, and hardened and granulation(keratinization)-ized it is produced. Then, CMC granulation-ized **above** is immersed in a lot of methanol for the purpose of drying, and after separating this methanol and drying, the granulation method of CMC of obtaining granular CMC which dries and serves as a granulation article is raised (US,2715124,B). However, in the above-mentioned granulation method, since gel CMC by spraying of water is uneven as

mentioned above, there is a problem that a granulation article with a particle size uniform as a product is not obtained. In order for gel CMC to stick and to grow up granular as mentioned above, the grain shape state is formed in the particles by which uneven shape was formed in the surface like konpeito, and, as a result, has the problem that mobility worsens. CMC of the degree of high ether substitution has high water solubility, and since it is easy to dissolve, adhesion to a device is strong, and since CMC sticks further and it does not become particles, there is a problem of being unable to carry out **granulation** -izing. The yield of product CMC is bad and the above-mentioned granulation method is a high cost at the whole.

0006 On the other hand, applicant of this application has proposed the method of carrying out spray drying and granulation-izing by making it flow down on a rotating disc, and making the solvent-water content slurry of CMC atomize previously (Tokuganhei6-215058). However, the CMC granulation article obtained by this method had low bulk density, and it turned out that it is not desirable in respect of the mobility of CMC, and miniaturization.

0007 It was made in view of such a situation, the particle diameter in the end of dried powder obtained is uniform, and generation of fines is controlled, the granulation method of CMC which can obtain CMC which was moreover excellent in handlability with high bulk density is carried out, and this invention sets offer of granular CMC as the purpose.

Means for Solving the Problem In order to attain the above-mentioned purpose, this invention makes the 1st gist a granulation method of CMC corned in liquid by cracking massive CMC in hydrous isopropyl alcohol, The whole contains not less than 80%, and particles of within the limits with a particle diameter of 149-2000 micrometers make the 2nd gist granular CMC whose bulk density is 0.4g/ml or more.

0009

Embodiment of the Invention Regardless of the degree of ether substitution, this invention from the degree of low ether substitution to the degree of high ether substitution, That is, even if it is water-soluble high CMC, in order to obtain the granulation article of quality CMC, it is the method of corning in liquid by cracking massive CMC in isopropyl alcohol (the following "IPA" is called) of water content. The system which consists of a hydrous IPA solution which contains massive CMC used as a raw material in the granulation in this liquid, The CMC granulation article with high bulk density (granular CMC) in which the content ratio of CMC is uniform particle diameter when the content ratio of IPA sets to 55 to 80% of the whole system and a moisture content ratio sets to 15 to 40% of the whole system 5 to 30% of the whole system, and generation of fines was controlled is obtained, and it is more effective. In the describing **above** granulation method in liquid, granular CMC which is uniform particle diameter and by which generation of fines of 75 micrometers or less was controlled for particle diameter is obtained by performing the Reynolds number (Re) which shows the flowability of the hydrous IPA solution system containing massive CMC under the conditions set or more to 1000. Therefore, when dissolving obtained granular CMC in water, the generation phenomenon of the unmixed-in lump of flour seen conventionally does not arise, but it dissolves in water promptly. As a result, the dissolving time at the time of use by a various application is shortened substantially, and since there is little dusting and mobility is still better, it excels also in handlability. At the time of granular CMC use, there is little generating of dust and it does not cause aggravation of work environment.

0010 And that to which not less than 80% of all the particles existed in within the limits with a particle diameter of 149-2000 micrometers, and bulk density was set **ml** in 0.4g /or more as granular CMC is excellent in mobility and solubility, and does not produce a raising dust problem, either, but, moreover, comes to have good handlability.

0011 Below, this invention is explained in detail.

0012 Although the degrees of average ether substitution are 0.4-3.0 and the target CMC can be applied to any CMC in the granulation method of CMC of this invention in that it is applicable also to CMC which has a binding property, In particular, in the high degree of average ether substitution of hydrophilic nature, CMC of 1.5-3.0 serves as a suitable object. Although sodium salt, potassium salt, ammonium salt, etc. are raised as a kind of alkali salt, it is usually sodium salt. CMC may be any of refining CMC obtained via the process of removing a byproduction salt using crude CMC and this crude CMC. Since the refining effects are inferior when demineralization refining is carried out after granulation-izing by this invention, it is preferred to use refining CMC.

0013 The granulation method of CMC of this invention is performed as follows, for example. That is, pulp which is a publicly known method is conventionally used for a raw material, neutralization processing is performed via an alkali-cellulose-ized reaction and an etherification reaction, and crude CMC is prepared by carrying out deliquoring filtration. On the other hand, prepare the partially aromatic solvent (hydrous IPA) which mixed IPA with water by the weight ratio within the limits of water/IPA=40 / 60 - 70/30, and the above-mentioned crude CMC is received, The weight of the above-mentioned crude CMC throws in 5-20 times the amount of this partially aromatic solvent, it stirs at 30-75 ** for 0.5 to 1 hour, and a demineralization step is performed (desalting process). And it deliquors with a centrifuge, a decanter, a screw-type continuation liquid separation machine, a filter, etc., and wet-like refining CMC is produced. Refining CMC produced here is mass material in which CMC solid content has elasticity **like the shape of the shape of agar to konnyaku which has a binding property by IPA consisting of 15 to 35% of presentation 45 to 65%** whose water is 20 to 40%. Thus, massive refining CMC obtained serves as high water content of 45 to 65% in the above-mentioned desalting process.

0014 Next, supply massive refining CMC and IPA (solvent) of 20% or less of water content to a predetermined mixer by the shape of wet acquired above, the moisture which remains in refining CMC is made to shift to it into the solvent which is IPA, and dehydrating treatment of refining CMC is performed. While performing this dehydrating treatment, by carrying out shuttlecock stirring and cracking massive refining CMC within a mixer, it cuts finely and the granulation in liquid is performed. Since said refining CMC is high water content, IPA of 20% or less of water content which uses by the above-mentioned granulation in liquid is used in order to adjust the water content to 5 to 30% of range. That is, IPA of 20% or less of water content is a useful solvent which can prevent re-binding at the time of a granulation, and can perform dehydrating treatment of refining CMC simultaneously. And since use of methanol and ethanol which are, other organic solvents, for example, lower alcohol, other than IPA, has the high water solubility and adhesion of CMC, it becomes impossible to dissolve and to perform the granulation in liquid, and it is not preferred. Since a bad smell cannot deaerate use of butanol easily strongly, it becomes expensive economically and is not preferred.

0015 Next, CMC corned among liquid removes a solvent with a centrifuge, then performs desiccation for 5 to 60 minutes at 60-110 ** using a fluidized-drying machine or a decompression type dryer. Thus, granular CMC is obtained.

0016 In the above-mentioned granulation in liquid, a mixer type mixer as shown in drawing 1 is raised as a mixer which supplies refining CMC and hydrous IPA. That is, the granulation in liquid is performed, rotating the knife type agitating blades 1, cutting massive refining CMC2 small, and adding rotation in this mixer, in the hydrous IPA solution 3 stirred with predetermined Reynolds number.

0017 In the above-mentioned granulation in liquid, it is preferred to perform the Reynolds number (Re) of the value which shows the flowability of hydrous IPA solution 3 system containing massive refining CMC2 by 1000 or more strong shuttlecock stirring. Reynolds number (Re) is 4000-30000 especially preferably. That is, granular CMC which is uniform particle diameter and by which generation of fines of 75 micrometers or less was controlled for particle diameter comes to be obtained by setting Reynolds number (Re) or more to 1000. The above-mentioned Reynolds number (Re) is computed by the formula (1) shown below.

0018

Equation 2 Re=(D-U-rho)/mu ... (1)

In a formula (1), as for shuttlecock peripheral speed (cm/sec) and rho of D, liquid density (g/cm^3) and mu are a shuttlecock diameter (cm) and U liquid viscosity (g/cm-sec).

0019 In the above-mentioned granulation in liquid, the number of rotations at the time of stirring is suitably set up within the limits of 100-2000 rpm according to the massive amount of refining CMC etc. to supply. As for the temperature of massive refining CMC to supply, mixing time is set as 10-50 ** in 5 to 30 minutes.

0020 And on the occasion of the granulation in liquid, supply of massive refining CMC may be supplied continuously and may be added to a batch process. It may add at once by a batch type, and the amount of supply is not limited.

0021 The granulation morphosis in such a granulation stage in liquid is explained in detail.

0022 That is, CMC after an ether reaction at the time of crude CMC manufacture is maintaining fibrous voice, and its density with bulky is low. A low water solvent usually performs demineralization refining in order to remove impurities, such as a byproduction salt contained in

this crude CMC. However, in this invention, it precedes supplying the granulation in liquid as mentioned above, and demineralization refining is carried out beforehand in high hydrous IPA whose water content is 40 to 70%. If a demineralization step is performed and filtrate separation is carried out in this high hydrous IPA, massive refining CMC of the shape of wet which contained moisture 45 to 65% will be obtained. At this process, since fibrous CMC contains moisture, it is welded and changes from the shape of agar to a massive konnyaku-like solid. The good state of dispersibility is held by stirring, without this solid dissolving in an IPA system solvent.

0023 Subsequently, after carrying out filtrate separation, for the purpose of drying of a granulation in liquid, and massive refining CMC, konnyaku-like massive refining CMC is supplied with 20% or less of hydrous IPA (solvent), rotates agitating blades and carries out stirring mixing here. And massive refining CMC is promptly cracked in liquid, and granulation cutting is carried out at particle diameter of about 1 mm. Here, drying by moisture shift to IPA advances, and, simultaneously with a crack, spherical particles are obtained by rotation.

0024 Thus, not less than 80% of the whole particle has a particle size within limits which are the particle diameter of 149-2000 micrometers, and obtained granular CMC does not contain fines with a particle diameter of 75 micrometers or less. Or even if it contains fines with a particle diameter of 75 micrometers or less, they are very little content of a grade which does not cause a soluble fall by an adverse effect by dust, and unmixed-in-lump-of-flour generation. Measurement of the above-mentioned particle diameter is measured by standard sieve (indicated to JIS Z8801), and can check the whole particle size distribution. Bulk density which is 0.4-0.8g/ml 0.4g/ml or more has **especially granular CMC that comprises such particle size distribution** still higher bulk density.

0025 Below, it combines with a comparative example and working example is described.

0026 First, three kinds of refining CMC which serves as a raw material on the occasion of a granulation of this invention and from which the degree of etherification (DS) differs were produced.

0027Production of refining CMC by solvent method** Stock pulpIt ground to 0.3 mm in diameter, having applied **the Kohjin Co., Ltd. make and NDSP (the sulfate method pulp)** to a grinder, and pulp of 5% of moisture content was obtained. then, the 30-l. reaction vessel made from SUS filled up with hydrous IPA which consists of IPA20 weight section (it omits the following "part") and two copies of water -- 0.8 copy of this crushed pulp -- in addition, it stirred for about 20 minutes at 20 **. And an alkali-cellulose-ized reaction was performed by in addition stirring 1.1 copies of flake articles of caustic alkali of sodium for 60 minutes at 30 ** for 5 minutes 100% to this. Next, 2.4 copies of monochloroacetic acid content IPA solutions were added over 10 minutes 50% to this. Then, after carrying out temperature up to 70 ** over about 30 minutes, again, it cooled to 30 ** and 1.1 copies of flake articles of caustic alkali of sodium were added over 5 minutes 100%, and 2nd addition was performed these 30 minutes afterward, having covered 2.4 copies of monochrome acetic acid content IPA solutions 50% for 10 minutes. Then, temperature up was carried out to 70 ** over 30 minutes, and an etherification reaction for 60 minutes was completed in this 70 ** state. After an etherification reaction, it cooled to 40 ** over 10 minutes, 0.3 copy of acetic acid solution was added 50%, and superfluous caustic alkali of sodium in a system was neutralized. And deliquoring filtration of this was carried out with a centrifuge, and 8.7 copies of crude CMC (sodium salt) of the degree 2.1 of etherification (DS) were produced. Water of CMC was **IPA of a presentation of this crude CMC** 28.5% 19.5% 52%.

0028 Six copies of water and a 60% hydrous solvent of four copies of IPA were added to one copy of above-mentioned crude CMC, stirring was performed to it for 60 minutes at 50 **, and a demineralization step was performed to it. Then, deliquoring filtration was carried out with a centrifuge and 0.8 copy of wet-like massive refining CMC (sodium salt) was obtained. Thus, obtained refining CMC was mass material which consists of 30% of solid content, 48% of water, and IPA22% of presentation, and has agar-like softness and brittleness.

0029Production of refining CMC by solvent method** Stock pulpIt ground to 0.3 mm in diameter, having applied **the Kohjin Co., Ltd. make and NDSP (the sulfate method pulp)** to a grinder, and pulp of 5% of moisture content was obtained. Then, 1.0 copy of this crushed pulp was added to the 30-l. reaction vessel made from SUS filled up with hydrous IPA which consists of 25 copies of IPA, and 2.5 copies of water, and it stirred for about 20 minutes at 20 **. And an alkali-cellulose-ized reaction was performed by in addition stirring 1.0 copy of caustic soda aqueous solution for 60 minutes at 30 ** for 5 minutes 40% to this. Next, 0.78 copy of monochloroacetic acid content IPA solution was added over 5 minutes 50% to this.

Then, temperature up was carried out to 70 ** over about 30 minutes. An etherification reaction for 90 minutes was performed in this 70 ** state. After an etherification reaction, it cooled to 40 ** over 10 minutes, 0.3 copy of acetic acid solution was added 50%, and superfluous caustic alkali of sodium in a system was neutralized. And deliquoring filtration of this was carried out with a centrifuge, and 2.5 copies of crude CMC (sodium salt) of the degree 0.6 of etherification (DS) were produced. Water of CMC was **IPA of a presentation of this crude CMC** 23.6% 11.4% 65%.

0030 Seven copies of water and a 70% hydrous solvent of three copies of IPA were added to one copy of above-mentioned crude CMC, stirring was performed to it for 30 minutes at 50 **, and a demineralization step was performed to it. Then, deliquoring filtration was carried out with a centrifuge and 2.3 copies of wet-like massive refining CMC (sodium salt) were obtained. Thus, obtained refining CMC was mass material which consists of 21% of solid content, 57% of water, and IPA22% of presentation, and has konnyaku-like elasticity and brittleness.

0031Production of refining CMC by solvent method** Stock pulpIt ground to 0.3 mm in diameter, having applied **the Kohjin Co., Ltd. make and NDSP (the sulfate method pulp)** to a grinder, and pulp of 5% of moisture content was obtained. Then, 1.0 copy of this crushed pulp was added to the 30-l. reaction vessel made from SUS filled up with hydrous IPA which consists of 25 copies of IPA, and 2.5 copies of water, and it stirred for about 20 minutes at 20 **. And an alkali-cellulose-ized reaction was performed by in addition stirring 2.7 copies of caustic soda aqueous solutions for 60 minutes at 30 ** for 5 minutes 40% to this. Next, 2.3 copies of monochloroacetic acid content IPA solutions were added over 5 minutes 50% to this. Then, temperature up was carried out to 70 ** over about 30 minutes. An etherification reaction for 90 minutes was performed in this 70 ** state. After an etherification reaction, it cooled to 40 ** over 10 minutes, 0.3 copy of acetic acid solution was added 50%, and superfluous caustic alkali of sodium in a system was neutralized. And deliquoring filtration of this was carried out with a centrifuge, and 5.6 copies of crude CMC (sodium salt) of the degree 1.5 of etherification (DS) were produced. Water of CMC was **IPA of a presentation of this crude CMC** 27.3% 14.7% 58%.

0032 Nine copies of water and a 60% hydrous solvent of six copies of IPA were added to one copy of above-mentioned crude CMC, stirring was performed to it for 30 minutes at 50 **, and a demineralization step was performed to it. Then, deliquoring filtration was carried out with a centrifuge and 1.4 copies of wet-like massive refining CMC (sodium salt) were obtained. Thus, obtained refining CMC was mass material which consists of 26% of solid content, 50% of water, and IPA24% of presentation, and has konnyaku-like elasticity and brittleness.

0033

Working example 1-15, comparison working example 1-6 A granulation by this invention was performed as follows using refining CMC** produced as mentioned above, **, and **. That is, one copy of solid content of massive refining CMC shown in the after-mentioned table 1 and this 8-times the amount 10% hydrous IPA were added to this using a large-sized mixer type mixer (made by a domestic centrifuge company) with a capacity of 10 l. first provided with the knife type agitating blades 1 as shown in drawing 1. Each content ratio of CMC of the whole system at this time, IPA, and moisture is shown in the after-mentioned table 1. And stirring was started at a room temperature (25 **), and it stirred by setting promptly Reynolds number which shows a flow of a solution as a value shown in the after-mentioned table 2. Granulation conditions by the above-mentioned mixer are shown in the after-mentioned table 2. The above-mentioned Reynolds number is the value computed by said formula (1), combines with the after-mentioned table 2 each value of each factor and shuttlecock diameter D (cm), the shuttlecock peripheral speed U (cm/sec), the liquid density rho (g/cm^3), and the liquid viscosity mu (g/cm-sec) needed for calculation of Reynolds number, and shows it.

0034

Table 1

For drawings please refer to the original document.

0035

Table 2

For drawings please refer to the original document.

0036 While massive refining CMC was small cracked in hydrous IPA liquid as a result of the above-mentioned stirring, since it rotated in liquid, granular CMC in which the granule child was formed spherically was obtained. The optical microscope photograph of granular CMC of one working example is shown in drawing 2. Thus, as for it, granular CMC obtained by the granulation in liquid turns out that the whole is approximately spherical. A lot of moisture contained in massive refining CMC shifted into IPA liquid on the occasion of the above-mentioned stirring. As a result, keratinization advanced with drying and, as for the CMC surface by which the granulation was carried out, solidification and the rise of density occurred. For this reason, CMC particles did not adhere easily to devices due to the shape of a pan pan, and the granulation article of good CMC was obtained. And the above-mentioned granulation process is performed in 5 to 20 minutes, and can be performed by any method of continuous system or a batch type.
0037 After the above-mentioned granulation, with the centrifuge, after carrying out deliquoring separation, the drying-by-warm-air machine performed desiccation of 2 hours at 100 ** warm air. Thus, granular CMC was obtained.
0038
Comparative examples 1-4 10 times the amount of hydrous methanol was added to this 20% using crude CMC** from which the degree of etherification produced in the manufacturing process of the above-mentioned refining CMC** - ** differs - **, demineralization washing was performed 2 times, and refining CMC was obtained. And in order to double a moisture content with above-mentioned working example for the above-mentioned refining CMC, spray water, it was made to increase and an 8-times the amount 10% hydrous methanol system performed granulation-ization after that on the conditions shown in the after-mentioned table 3 using the same mixer as above-mentioned working example (comparative examples 1-3). On the other hand, after hot air drying equipment performed desiccation of 100 **x 2 hours, the shock type pulverizing mill (made by Hosokawa Micron CORP.) ground deliquored refining CMC, and granular CMC was obtained (comparative example 4).
0039
Table 3

For drawings please refer to the original document.

0040
Conventional example 15 times the amount of hydrous methanol was added to this 20% using crude CMC** of the degree 1.5 of etherification produced in the manufacturing process of the above-mentioned refining CMC**, demineralization washing was performed 2 times, and refining CMC was obtained (CMC in refining CMC: 65%, 5% of moisture, methanol 30%). And in order to double a moisture content with above-mentioned working example for this refining CMC, sprayed water, it was made to increase to 50% of a moisture content, and CMC which melted a part of fibrous CMC, and hardened and (keratinization) corned it was produced. Then, for the purpose of drying, 10-times the amount 100%IPA was added to this CMC, and after separating the solvent and drying, granular CMC was obtained by performing 60 ** hot-air-drying processing. The optical microscope photograph of granular CMC of the obtained conventional example article is shown in drawing 3. Thus, as for obtained granular CMC, uneven shape was formed in the whole surface **like konpeito** the shape of whose a grain shape is.
0041 Thus, in each granular CMC of elegance, the yield and water content of the state of granular CMC corned under the granulation in liquid and obtained granular CMC are shown in the after-mentioned Table 4 and 5 the obtained working example article, a comparison working example article, a comparative example article, and conventionally. The yield of the above-mentioned granular CMC is computed by the following formula.
0042
Equation 3 Yield (%) = **(purity of granular CMC) /(purity of refining CMC)** x100 **0043**The bulk density of each obtained granular CMC, the content ratio of the particles of within the limits with a particle diameter **to the whole granular CMC** of 149-2000 micrometers, and the content ratio of a particle with a particle diameter of 75 micrometers or less are measured by a standard sieve (JIS Z8801), and the result is shown in the following Table 4 and 5.
0044
Table 4

For drawings please refer to the original document.

0045
Table 5

For drawings please refer to the original document.

0046 The chart figure showing the particle size distribution by measurement by the standard sieve (JIS Z8801) of five working example (the degree 1.5 of ether substitution) and nine working example (the degree 2.1 of ether substitution) is shown in drawing 4 (five working example) and drawing 5 (nine working example) out of measurement of the above-mentioned call counter, respectively, The numerical value acquired by the measurement is shown in the following Table 6 (five working example) and 7 (nine working example). 381.52 micrometers and the mean particle diameter of nine working example of the mean particle diameter of five working example by the above-mentioned call counter measurement were 814.41 micrometers, standard deviation was 597.28 micrometers, and standard deviation was 267.09 micrometers.
0047
Table 6

For drawings please refer to the original document.

0048
Table 7

For drawings please refer to the original document.

0049 The solubility of each CMC and the dissolution rate, the dusting characteristics, the amount of raising dust, adhesion, and mobility which were obtained were measured and evaluated in accordance with the following method. The result is shown in the after-mentioned table 8.
0050The solubility and the dissolution rate of granular CMC 800 ml of water was put into a 1000-ml beaker, and 8 g (1% concentration) of CMC samples were added into this. As a result, the thing of x and the above-mentioned middle evaluation of what became a letter of distribution immediately and was able to perform the aqueous solution quickly was displayed as **. **O and the thing which the unmixed-in-lump-of-flour-like lump was formed immediately and the aqueous solution took the long time** Time until it stirs lightly and a CMC sample dissolves was measured.
0051Dusting characteristics of CMC 1 / 2 capacity restoration of the CMC sample were carried out at the 100-ml screw pipe, and this was stirred up and down. As a result, the thing of x and the above-mentioned middle evaluation of a thing without ****** of fines was displayed as **. **what has many ****** of O and fines**
0052The amount of raising dust of CMC After putting 10g of CMC samples into the 300-ml Mayer flask and shaking at 10 cm of upper and lower sides 3 times, the floating CMC dust was absorbed by the suction collector which set the extraction thimble, and the amount of raising dust was measured.
0053Adhesion of CMC After filling up a 1-l. polyethylene bag with 200g of CMC samples and neglecting it on the 1st, the bag mouth was placed upside down, the natural fall of the CMC sample was carried out, and it took out from the inside of a bag. And the CMC powder quantity which adhered and remained in the bag was measured, and deposit efficiency was computed.
0054Mobility of CMC From the 20-cm-high place, it let the funnel whose inside diameter of the exit lower part is 10 mm pass, and the natural fall of 50 g of the CMC samples was carried out on the glass plate. And the diameter of CMC which fell and spread in the round form on the glass plate was measured. It can be said that what has a large diameter naturally has high mobility.
0055
Table 8

For drawings please refer to the original document.

0056 From the result of the above-mentioned table 8, a comparative example article is inferior to solubility, and its amount of raising dust is also high. It turns out that it is inferior also to mobility by high adhesion. On the other hand, the comparison working example article excels the comparative example article in each characteristic. The working example article produced by specifying Reynolds number or more as 1000 is dramatically excellent in solubility and mobility.
And it is clear that there are not the amount of raising dust and coating weight, and the good result was obtained in each characteristic.

Brief Description of the Drawings

Drawing 1 It is a mimetic diagram showing the composition of the mixer type mixer used with the granulation method of CMC of this invention.
Drawing 2 It is an optical microscope photograph 50 times the magnification of this which shows the particulate structure of granular CMC of one working example acquired by the granulation in liquid.
Drawing 3 It is an optical microscope photograph 50 times the magnification of this which shows the particulate structure of granular CMC obtained by the conventional process.
Drawing 4 It is a chart figure showing an example of the result of having measured the particle size distribution of granular CMC which is five working example (the degree 1.5 of ether substitution) by the standard sieve (JIS Z8801).
Drawing 5 It is a chart figure showing an example of the result of having measured the particle size distribution of granular CMC which is nine working example (the degree 2.1 of ether substitution) with the call counter.

Drawing 1

For drawings please refer to the original document.

Drawing 2

For drawings please refer to the original document.

Drawing 3

For drawings please refer to the original document.

Drawing 4

For drawings please refer to the original document.

Drawing 5

For drawings please refer to the original document.

---------- **Written Amendment**
Filing date Heisei 7(1995) August 11
Amendment 1
Document to be AmendedDRAWINGS
Item(s) to be Amended Drawing 2
Method of Amendment Change
Proposed Amendment
Drawing 2

For drawings please refer to the original document.

Amendment 2
Document to be AmendedDRAWINGS
Item(s) to be Amended Drawing 3
Method of Amendment Change
Proposed Amendment
Drawing 3

For drawings please refer to the original document.